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MEASUREMENTS OF NITRIC OXIDE ION VIBRATIONAL ABSORPTION COEFFICIENT AND VIBRATIONAL TRANSFER TO N_2

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Line Locations
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Vibrational Transfer Rates

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The line locations absorption coefficient and quenching of $NO^+(v)$ vibration in the v = 1 and v = 2 by N_2 have been experimentally determined. Integrated absorption coefficients corrected for a thermal equilibrium distribution are: S_{lu}(1 - 2 P15) = 420 cm^{-2}/atm^{-1} at 2291.63 cm^{-1} and S_{lu}(2 - 3 R2) = 653 cm^{-2}/atm^{-1} at 2286.72 cm^{-1}. Quenching of $NO^+(v)$ by N_2 at 297°K was found to give rates of $k = 2.4 \times 10^{-12}$ cm^3/sec^{-1} for $NO^+(v = 2)$ and $k = 3.0 \times 10^{-12}$ cm^{-3}/atm^{-1} for $NO^+(v = 1)$. An estimate of 3×10^{-13} cm^3/sec^{-1} was found for quenching of $NO^+(v = 2)$ by NO at 297°K.

Times 10 to the -13th power cu.cm.

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MEASUREMENTS OF NITRIC OXIDE ION VIBRATIONAL ABSORPTION COEFFICIENT AND VIBRATIONAL TRANSFER TO N_2

1. INTRODUCTION

Interest in NO^+ vibrational band intensities stems from its relatively large population in electron disturbed atmospheres. The nitric oxide ion is the most abundant ion in auroral and other electron excited events. Principal sources of NO^+ in the upper atmosphere are the ion exchange reactions $N_2^+ + O \rightarrow NO^+ + N$ and $O^+ + N_2 \rightarrow NO^+ + N$ and the charge exchange reactions of N_2^+, N^+, O^+, and O_2^+ with natural atmospheric NO. Since all of these reactions are exothermic enough to excite several vibrational levels in NO^+, $NO^+(v)$ may provide an important source of infrared emission in the electron disturbed upper atmosphere.

Radiation from vibration of $NO^+ (X\ ^1\Sigma^+)$ is in the 4.3 μm band, coincident with the $CO_2(\nu_3)$ emission. While $CO_2(\nu_3)$ emissions dominate in the undisturbed atmosphere, self-absorption by cold CO_2 limits its emission signal to the wings of the CO_2 band. For any sensor system designed to operate in these regions, the effects of $NO^+(v)$ radiation on these systems may be of great importance. Because of the intense $CO_2(\nu_3)$ emission, $NO^+(v)$ radiation has not been detected during auroral events except perhaps to explain very prompt emission during an IBC III aurora.[1,2] The only unambiguous NO^+ emission in the upper atmosphere was observed during a nuclear test in 1962.[3]

While the radiative lifetime of NO^+ has never been measured, Stair and Gauvin[3] have inferred an integrated absorption coefficient of $S_{lu} = 500\ cm^{-2}\ atm^{-1}$ from the 1962 nuclear observations past the CO_2 blue spike region. This value was obtained through various assumptions of vibrational equilibrium and atmospheric transmission. The only other information of the vibrational lifetime of NO^+ has been from ab initio

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(1) J. Ulwick, ICECAP 1975 Data, presented at DNA HAES Meeting (June 1977).

(2) J. Kumer, "Analysis of 4.3μ ICECAP Data," AFCRL-TR-740334 (July 1974).

(3) A. T. Stair and H. P. Gauvin, Aurora and Airglow, B. M. McCormac, Ed. Reinhold Press (1967).

calculations by F. Billingsley[4] and H. H. Michels.[5] Billingsley obtained a ground state integrated absorption coefficient of S_{lu} = 88.9 cm^{-2} atm^{-1} using a multi-configuration self-consistent field (MC SCF) approach. Similarly, Michels arrived at a calculated value of S_{lu} = 168 cm^{-2} atm^{-1}.

The purpose of this study is to measure the radiative lifetime of NO^+ vibrational bands in the $X\,^1\Sigma^+$ ground state and to measure the transfer of vibration between NO^+ and N_2 whose vibrational energies are in close resonance.

This paper describes the experimental determination of NO^+ $\Delta v = 1$ integrated absorption coefficient in the v = 0, v = 1, and v = 2 levels, and quenching of the v = 1, v = 2, and v = 3 levels by N_2. The experimental arrangement is presented in Section 2. Results of the experimental determination of the NO^+ absorption coefficient are presented in Section 3. In Section 4, we discuss the experimental determination of the quenching of $NO^+(v)$ by N_2. A discussion of our results is presented in Section 5.

[4] F. Billingsley, II, Chem. Phys. Lett 23 160 (1973).

[5] H. H. Michels, "Air Molecular Computation Study," AFGL TR-77-0032 (1977).

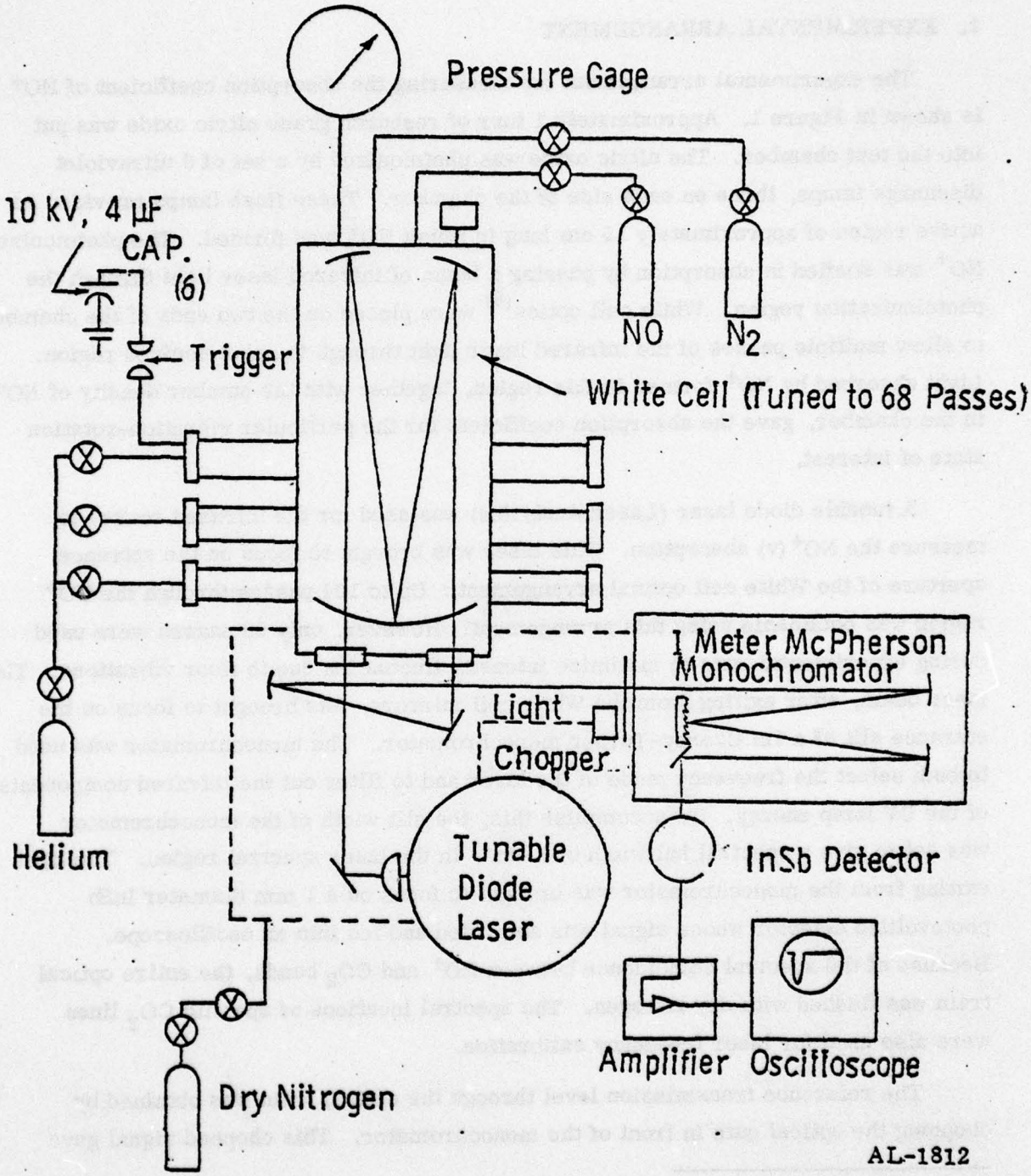

Figure 1. Schematic Diagram of Experimental Arrangement

2. EXPERIMENTAL ARRANGEMENT

The experimental arrangement for measuring the absorption coefficient of NO^+ is shown in Figure 1. Approximately 1 torr of research grade nitric oxide was put into the test chamber. The nitric oxide was photoionized by a set of 6 ultraviolet discharge lamps, three on each side of the chamber. These flash lamps provided an active region of approximately 15 cm long in which NO^+ was formed. The photoionized NO^+ was studied in absorption by passing a beam of infrared laser light through the photoionization region. White cell optics[6] were placed on the two ends of the chamber to allow multiple passes of the infrared laser light through the photoionized region. Light absorbed by NO^+ formed in this region, together with the number density of NO^+ in the chamber, gave the absorption coefficient for the particular vibration-rotation state of interest.

A tunable diode laser (Laser Analytics) was used for the infrared source to measure the $NO^+(v)$ absorption. This laser was brought to focus on the entrance aperture of the White cell optical arrangement. Up to 104 passes through the NO^+ region was obtainable using this arrangement. However, only 68 passes were used during experimental runs to minimize intensity fluctuation due to floor vibrations. The laser beam, after exiting from the White cell mirrors, was brought to focus on the entrance slit of a 1m Czerny-Turner monochromator. The monochromator was used to both select the frequency mode of the laser and to filter out the infrared components of the UV lamp energy. To accomplish this, the slit width of the monochromator was set to give a spectral halfwidth of 1 cm^{-1} in the laser spectral region. The light exiting from the monochromator was brought to focus on a 1 mm diameter InSb photovoltiac detector whose signal was amplified and fed into an oscilloscope. Because of the spectral coincidence between NO^+ and CO_2 bands, the entire optical train was flushed with dry nitrogen. The spectral locations of specific CO_2 lines were also used for laser frequency calibration.

The reference transmission level through the optical train was obtained by chopping the optical path in front of the monochromator. This chopped signal gave

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[6] J.U. White, J. Opt. Soc. Am. 32, 285 (1942).

the system transmission in the absence of NO^+. The chopper was then turned off in the open position during the operation of the photoionizing lamps. The ratio of laser intensity transmitted after the formation of NO^+ to that before formation of NO^+, thus provided the spectral extinction rate as a function of the laser wavelength and time. Since the halfwidth of the laser line ($\sim 10^{-4}$ cm^{-1}) was much narrower than the Doppler width of the absorbing line ($\sim 5 \times 10^{-3}$ cm^{-1}), the integrated absorption coefficient of the $NO^+(v, J)$ transition could be measured by tuning the laser across several points of the absorption line.

A typical calibration curve for the tunable diode laser is shown in Figure 2. Because of the temperature cycling of the diode from room temperature to 10^oK as the laser Dewar was filled, the actual tuning range varied from day to day. However, the laser usually tuned over at least one vibration rotation line per temperature cycling. Note that the tuning slope of the laser varied with current. The actual tuning slope for the particular wavelength of interest was obtained for each run in order to obtain the absorption halfwidth as well as location of the absorbing $NO^+(v)$ line.

The ultraviolet lamps used in this apparatus were of a modified Lyman configuration providing a 29000^oK blackbody continuum radiation when discharged.[7, 8] Helium gas was passed through the lamps at approximately 3 torr pressure. All 6 lamps were discharged simultaneously by triggering 6 spark gaps in parallel, each connecting 1 flash lamp to a high speed capacitor charged to 10 kv at 4 μF. The time synchronization between lamp discharges was within 1 and 2 μs in all experimental runs.

The output from each lamp was monitored by an ionization detector placed behind the region monitored by the White cell optics in the absorption chamber. The purpose of these ion detectors was to ensure that sufficient ion pairs were produced during each experimental run. The actual ion density, as a function of time, was determined separately. Each ionization detector consisted of a pair of parallel

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[7] T. Lyman, Science 64, 89 (1926).

[8] J. F. Holzrichter and J. L. Emmett, Appl. Opt. 8, 1459 (1969).

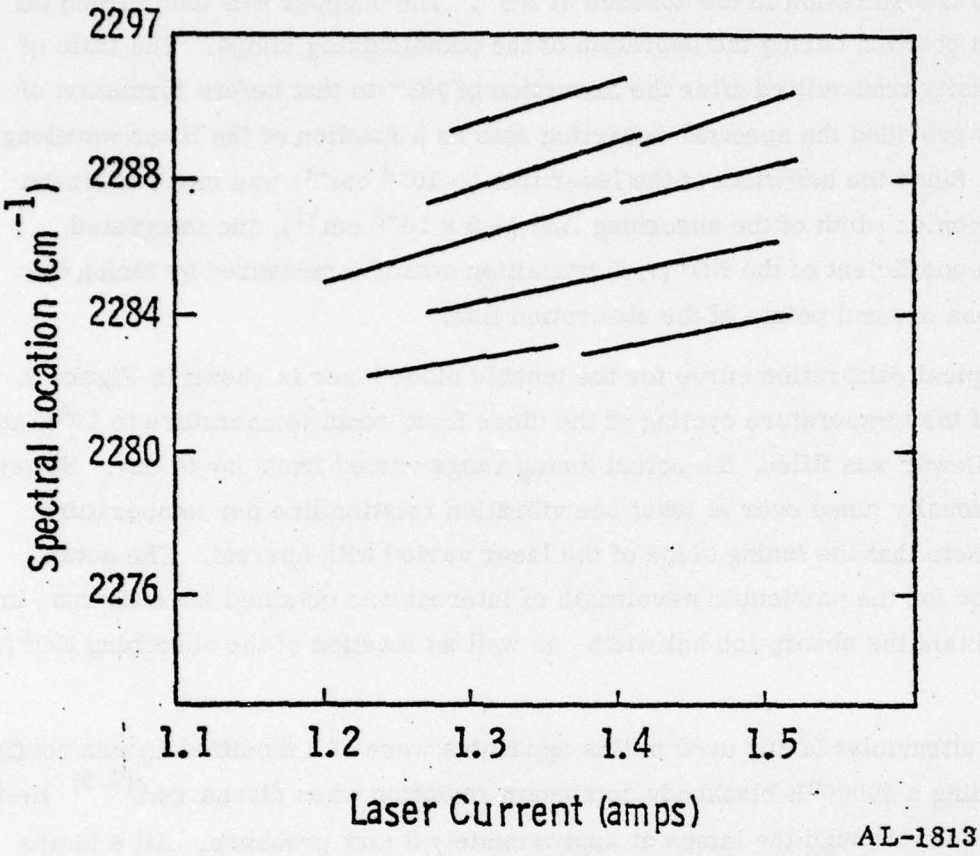

Figure 2. Typical Laser Turning Curves From Diode Laser. Relative Laser Intensities Varied With Time and Laser Current.

plates across which was put a 300V potential. Electrons produced in the photoionization would be attracted to the anode plate causing current to flow. The cathode and anode were shielded from UV light through a pair of guard rings placed at ground potential. The potential difference between the anode and ground was measured across 330Ω resistor. The change in potential was then proportional to the current through the anode, hence, the number of photoelectrons produced. Using this measurement scheme, it was established that more than 10^{13} ions pairs per cm^3 were produced per flash along the lamp axis within the absorption cell when the cell was filled to 1 torr with NO.

In order to obtain the vibrational transfer cross section from $NO^+(v)$ to N_2, provision was also made to bleed a partial pressure of N_2 into the absorption chamber. The pressure in the cell was monitored by a baratron pressure gage, reading from 0 to 10 torr with a least count of 10^{-4} torr. The partial pressure of N_2 was raised until $NO^+(v)$ transfer to N_2 caused a more rapid decrease in the extinction of diode laser signal in the $v = 1$ and higher vibrational states than would be attributable to dissociative recombination. The quenching of laser light extinction by N_2, as a function of N_2 pressure, thus gave a transfer rate of $NO^+(v, J)$ to N_2.

The absorption of laser light by NO^+, formed during photoionization typically, gave a signal as shown in Figure 3. This trace represents the ac component of laser light falling on the detector. Discharge noise caused a large positive spike which triggered the oscilloscope. NO^+ formed by photoionization absorbed the laser emission giving the drop in total intensity. Dissociative recombination, together with quenching of the vibration rotation line by NO and, as in Figure 3, 1 torr of N_2, caused the signal level to return to its dc level. The noise at late times was typical of the detector noise within the system even when no laser signal was present.

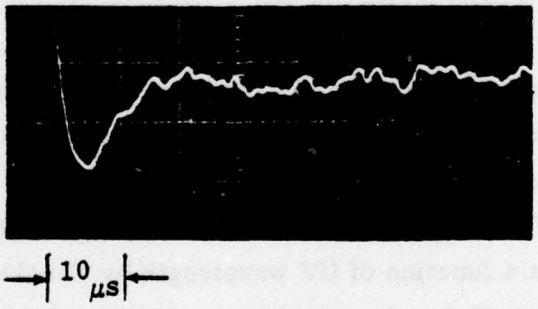

Figure 3. Typical Oscilloscope Trace of The ac Component of Transmitted Laser Light. Horizontal Scale Represents 10 μs/div, Vertical 20 mV/div. The Test Chamber Was Filled With 1 torr of NO and 1 torr of N_2. The Laser Line Was Tuned to Approximately the Peak of The NO^+ $v = 2$ to $v = 3$, R2 Absorption Line.

3. MEASUREMENT OF NO^+ ABSORPTION COEFFICIENT

The absorption coefficient for the $\Delta v = 1$ vibrational transition in $NO^+(X\ ^1\Sigma^+)$ ground electronic level was obtained by measuring the time varying extinction described above. When the laser wavelength was coincident with the $NO^+(v)$ line, some extinction of laser radiation would result after the flash lamps were fired as seen in Figure 3. This time varying extinction was a measure of both the absorption coefficient and the number densities of the states involved in the transition. The populations of the two states involved in the transition was determined from the dissociative recombination rate of NO^+ and the relative populations of NO^+ in the various vibrational levels. Rotational equilibrium was assumed throughout the experiment. Vibrational equilibrium, on the other hand, was assumed slow in the time scale of the experiment.

The relative concentrations of the lower and upper vibrational states in the transition were determined through fitting the measured window transmission with a predicted spectrum from the UV lamps and previously measured spectral data on photoionization.[9] Since MgF_2 windows were used in front of the UV lamps, the bandpass through which photoionization could occur was limited from 1130 to 1337A. Fortunately, there have been many studies in the threshold region of the NO photoionization.[9,10,11] From the measured photoionization efficiency in this region, the measured window transmission, and assuming a lamp output of a 30000°K blackbody continuum, the relative photoionization was arrived at as a function of wavelength and plotted in Figure 4. Also plotted, are the effects of UV lamp blackbody temperature of 40000 and 20000°K. The vibrational partition, using this technique and the vibrational partitions measured as a function of UV wavelength by Tanaka, et al.,[9] gave 0.29, 0.39, 0.22, 0.08, and 0.02 for the relative populations of the v = 0 through v = 4 levels, respectively,

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[9] K. Tanaka, K. Honma, I. Loyano, and I. Tanaka, J. Chem. Phys. 60, 3347 (1974).

[10] P. C. Killgoar, Jr., G. E. Leroi, J. Berkowitz, and W. A. Chupka, J. Chem. Phys. 58, 803 (1973).

[11] K. Watanabe, F. M. Matsunaga, and H. Sakai, Appl. Opt. 6, 391 (1967).

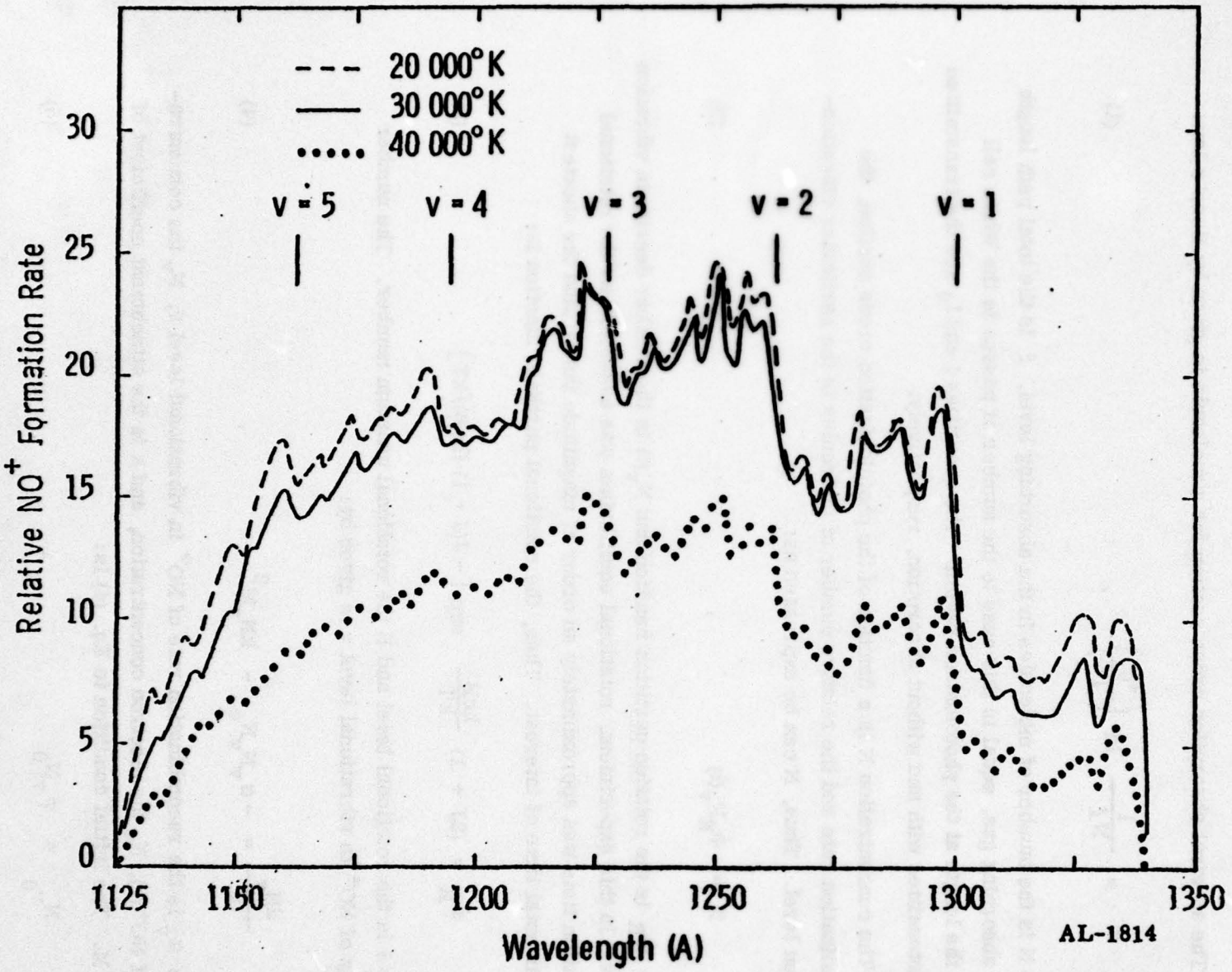

Figure 4. NO Photoionization Efficiency Through MgF_2 Windows As a Function of Lamp Output Temperature. Threshold Energy for Exciting v = 1 Through v = 5 Vibrational Levels of NO^+ Are Shown. Relative NO^+ (v) Concentrations Obtained by Folding This Spectrum With Vibrational Partitions Given in Ref. 9.

The spectral absorption cross section for a molecule is given by Beer's law:

$$\sigma = \frac{1}{N\ell} \ln\left(\frac{I_0}{I}\right) \quad , \tag{1}$$

where N is the number of molecules in the absorbing level, ℓ is the total path length in the absorbing gas, equal in this case to the number of passes in the White cell times the length at the photoionized region. The quantities I and I_0 are the transmitted light intensities with and without absorption, respectively.

The concentration N is a function of the photoionization cross section, the recombination rate and the relative number of molecules in the particular vibration-rotation level. Thus, N can be expressed as:

$$N = \phi_R N_v(t) \quad , \tag{2}$$

where ϕ_R is the rotation partition function and $N_v(t)$ is the number density in vibration level v. In this experiment, rotational equilibrium was assumed, as the rotational relaxation time was approximately an order of magnitude faster than the shortest experimental time of interest. Thus, the rotational partition function is:

$$\phi_R = (2J + 1) \frac{hcB}{kT} \exp\left[-J(J + 1)\,(hcB)/kT\right] \quad , \tag{3}$$

where J is the rotational level and B the rotational quantum number. The number density of NO^+ in vibrational level v is given by:

$$\frac{dN_v}{dt} = -\alpha_v N_v N_e - kN_v M^2 \quad , \tag{4}$$

where α_v is the recombination rate of NO^+ in vibrational level v, N_v the concentration of $NO^+(v)$, N_e the electron concentration, and k is the attachment coefficient of N_v to M. The initial condition to Eq. (4) is:

$$N_{v_0} = \phi_v N_0 \quad , \tag{5}$$

where ϕ_v is the relative production rate of NO^+ in vibrational level v and N_0 is the total number density of NO^+ produced. By operating at low enough NO pressures so that the second term on the right-hand side of Eq. (4) may be neglected, and assuming α_v is independent of v, the concentration of NO^+ in the vibrational level v, and rotational level J, is:

$$N = \phi_R \phi_v \frac{N_0}{1 + N_0 \alpha_v t} \quad . \tag{6}$$

The total absorption by $NO^+(v, J)$, from Eq. (1), must be time dependent, such that, $\ln (I_0/I)$ must vary as 1/N. During the experiment, the ratio $(I_0 - I)/I_0$ was at all times less than 10^{-2}. Thus, $\ln (I_0/I)$ may be substituted by the expansion giving:

$$\ln \frac{I_0}{I} \simeq \frac{I_0 - I}{I_0} \equiv \frac{\Delta I}{I_0} \quad . \tag{7}$$

Note that the choice of t = 0 in Eq. (6) can be rather arbitrary, provided that there is no production term in Eq. (4). Since both $\Delta I/I_0$ and 1/N must have the same time dependence, the concentration, N, at any time, t_0, may be calculated by the relation

$$\frac{\Delta I(t_0)}{\Delta I(t)} = 1 + N(t_0)\ \alpha_v (t - t_0) \quad . \tag{8}$$

Here, it may be seen that $N(t_0)$ is not a function of the total extinction but only of the rate of change of extinction with time. Knowing the recombination rate coefficient and the time varying extinction signal, measured as the ac component to the total laser light transmitted through the optical system, thus gives a number density. It is assumed here, that no other quenching of $NO^+(v)$ is occurring simultaneously.

The tuning range of the laser allowed the study of several vibration rotation lines in NO^+. These lines are listed in Table 1. While there were laser lines to study all of the $NO^+(v)$ lines listed, measurements of the integrated absorption coefficient were made only of the P5 line of the v = 1 to v = 2 transition and the R2 line of the

TABLE 1. $NO^+(v)$ ROTATION LINES WITHIN LASER BANDPASS

Vibrational Transition		Calculated Location (cm^{-1})	Measured Location (cm^{-1})	Cross Section at Peak (cm^{-2})
v = 0 to 1	P12	2294.1		
	P13	2289.7	2289.76	5×10^{-16} (±2x)
	P14	2285.2		
	P15	2280.7		
v = 1 to v = 2	P5	2291.6	2291.63	$1.5 \pm 0.4 \times 10^{-15}$
	P6	2287.4	2287.45	$1.3 \pm 0.5 \times 10^{-15}$
	P7	2283.3	2283.35	
v = 2 to v = 3	R1	2282.8		
	R2	2286.7	2286.72	$2.2 \pm 0.3 \times 10^{-15}$
	R3	2290.4		
	R4	2294.4		
v = 3 to v = 4	R10	2282.7		
	R11	2286.1	2286.30	$2 \pm 1.5 \times 10^{-15}$
	R12	2289.5		
v = 4 to v = 5	R20	2281.9		
	R21	2284.9		
	R22	2287.8		

v = 2 to v = 3 transition. In addition, spectral absorption coefficients were obtained for the R11 line of the v = 3 to v = 4 transition and the P13 line of the v = 0 to v = 1 transition. Unfortunately, due to the low signal-to-noise of these latter two measurements, sufficient data was not available to obtain a meaningful integrated absorption coefficient from these two transitions. Similarly, the P6 line of the v = 1 to v = 2 transition was measured using a rather weak laser line, which was near the mode shifting threshold of the laser. Because of the uncertainty introduced by the laser shift, the integrated absorption coefficient was not obtainable using this transition.

The locations of the lines of interest were found by measuring the locations of known CO_2 lines[12] and interpolating across the tuning range to obtain the precise location of the NO^+. In this way, the P5 line of NO^+ (v = 1 to v = 2) was found to be at 2291.63 cm^{-1}, while R2 line of the v = 2 to v = 3 transition was found to lie at 2286.72 cm^{-1}. These measured values were within 0.1 cm^{-1} of measurements by Miescher,[13] who deduced the vibrational structure from electronic transitions of NO^+.

The concentration of NO^+ was calculated using a least squares fit of the data to Eq. (8) assuming $\alpha_v = 4.3 \times 10^{-7}\ cm^3\ sec^{-1}\ (T_e/300)^{-0.37}$.[14] The relative population of the lower state involved in the absorption transition was then given by Eq. (6). Using this technique, the spectral absorption coefficient was obtained as a function of laser current from Eq. (1). The spectral absorption cross section for the P5 line of the v = 1 to v = 2 and the R2 line of the v = 2 to v = 3 transitions are shown in Figures 5 and 6, respectively. The laser current, plotted on the abscissa of these two figures, was fit to the tuning slope shown in Figure 2 to obtain the linewidth. The laser tuning rate was 7 cm^{-1} per ampere, giving an absorption halfwidth of 0.0056 cm^{-1} approximately the Doppler width for NO^+ at 300°K. The integrated absorption coefficient was thus computed using a 300°K Doppler profile.

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[12] R. Oberly, K.N. Rao, Y.H. Hahn, and T.K. McCubbin, Jr., J. Mol. Spect. 25, 138 (1968).

[13] E. Miescher, Helv. Physica, Acta 29, 135 (1956).

[14] C.M. Huang, M.A. Biondi, and R. Johnsen, Phys. Rev. A11, 901 (1975).

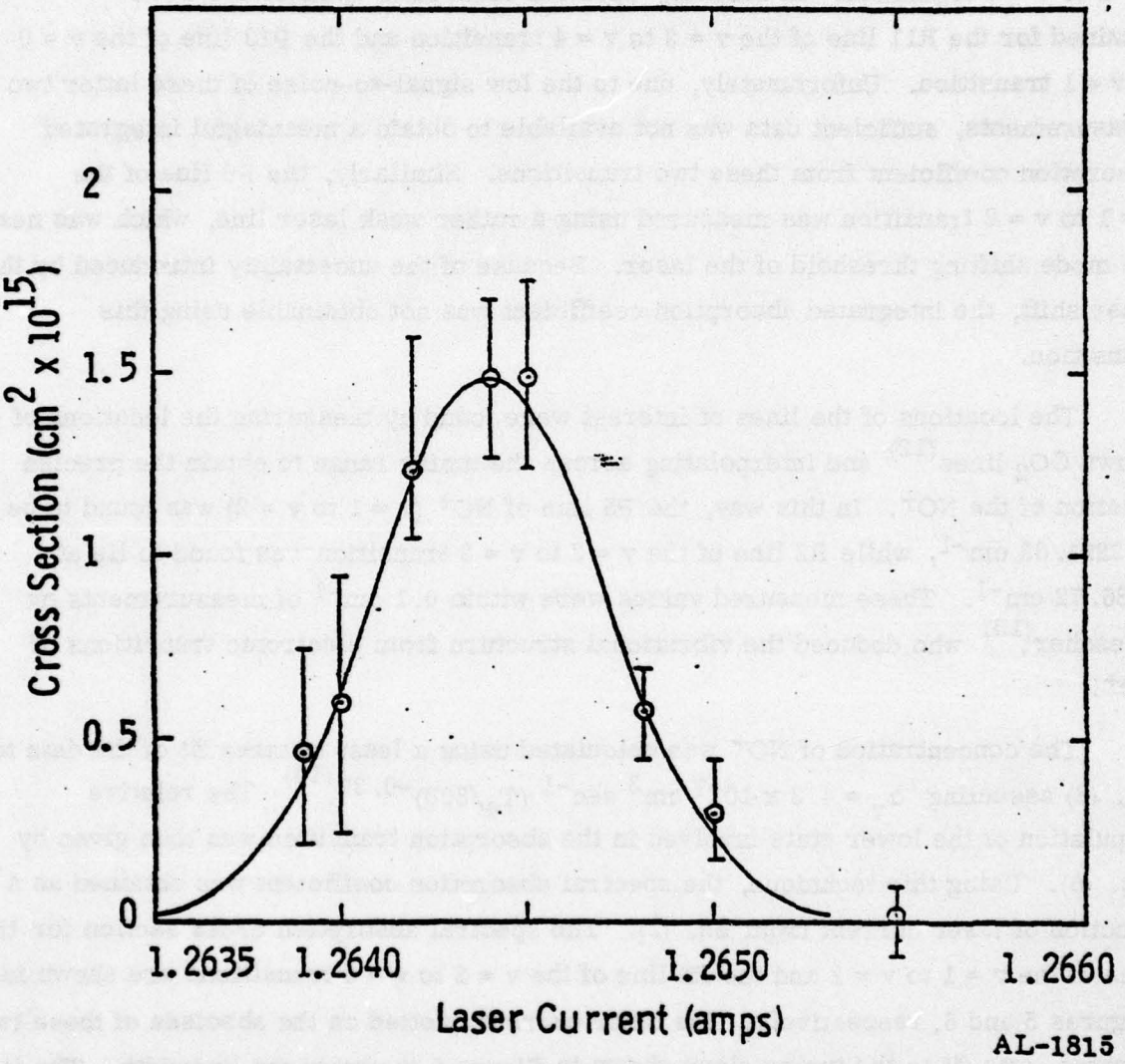

Figure 5. Spectral Absorption Cross Section of P5 Line of v = 1 to v = 2 Level Band of NO^+. Line Center at 2291.63 ± 0.01 cm^{-1}, Tuning Rate of 7 cm^{-1}/amp.

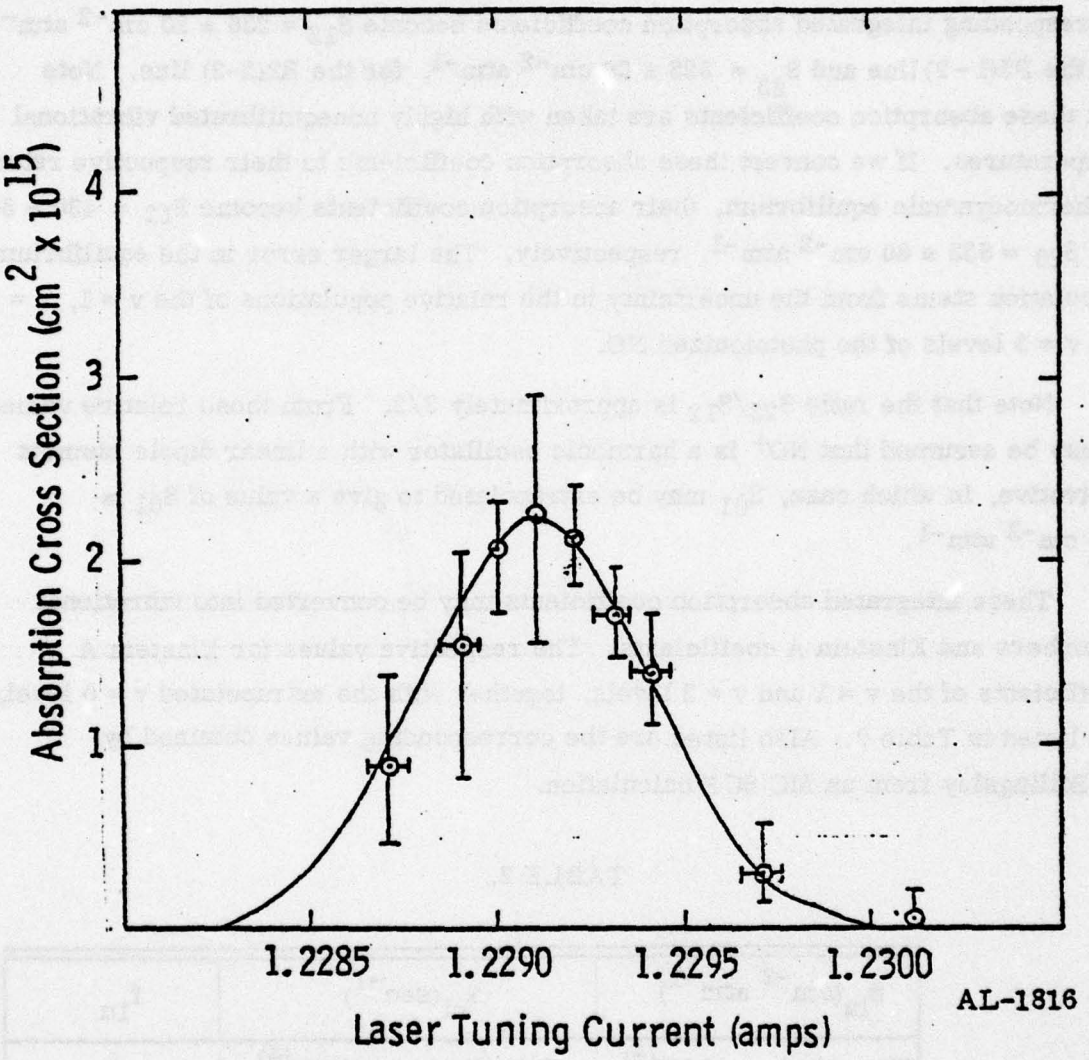

Figure 6. Spectral Absorption Cross Section of The R2 Line In The v = 2 to v = 3 Band. Tuning Rate Equals 7 cm^{-1}/amp. Line Location at 2286.72 ± 0.01 cm^{-1}.

The absorption cross section was obtained by assuming that the relative population in the v = 1, v = 2, and v = 3 levels were 0.39, 0.22, and 0.08, respectively. The corresponding integrated absorption coefficients become S_{12} = 236 ± 20 cm^{-2} atm^{-1} for the P5(1-2) line and S_{23} = 325 ± 20 cm^{-2} atm^{-1}, for the R2(2-3) line. Note that these absorption coefficients are taken with highly nonequilibrated vibrational temperatures. If we convert these absorption coefficients to their respective rates at thermodynamic equilibrium, their absorption coefficients become S_{12} = 430 ± 50 and S_{23} = 653 ± 80 cm^{-2} atm^{-1}, respectively. The larger error in the equilibrium calculation stems from the uncertainty in the relative populations of the v = 1, v = 2, and v = 3 levels of the photoionized NO.

Note that the ratio S_{23}/S_{12} is approximately 3/2. From these relative values, it may be assumed that NO^+ is a harmonic oscillator with a linear dipole moment derivative, in which case, S_{01} may be extrapolated to give a value of $S_{01} \cong$ 215 cm^{-2} atm^{-1}.

These integrated absorption coefficients may be converted into vibrational f-numbers and Einstein A coefficients. The respective values for Einstein A coefficients of the v = 1 and v = 2 levels, together with the extrapolated v = 0 level, are listed in Table 2. Also listed are the corresponding values obtained by F. Billingsley from an MC SCF calculation.

TABLE 2.

	S_{lu}(cm^{-2} atm^{-1})		A_{ul}(sec^{-1})		f_{lu}
	This Work	SCF[4]	This Work	SCF[4]	
v = 2 to 3	653		95.8		2.7×10^{-5}
v = 1 to 2	430	176	63.4	26.2	1.8×10^{-5}
v = 0 to 1	215	88.9	31.5	13.6	9×10^{-6}

4. MEASUREMENT OF THE NO^+ VIBRATIONAL EXCHANGE WITH N_2

The transfer rate of vibration from the v = 1 and v = 2 levels of NO^+ to N_2 was measured by observing absorption in the P5 and R2 absorption lines of these two vibrational levels, respectively. By using the absorption of infrared light as a monitor, the relative populations of the v = 0, v = 1, v = 2, and v = 3 levels were monitored as a function of N_2 pressure. Since the extinction of laser light is directly proportional to the population of the absorbing level, the time varying signal is a measure of vibrational transfer from NO^+. The disappearance of NO^+ (v) is from the following processes:

(R1) $NO^+(v) + e \rightarrow N + O$	$\alpha = 4.3 \times 10^{-7}(T_e/300)^{-0.37}$ $cm^3\ sec^{-1}$ (14)
(R2) $NO^+(v) + NO + NO \rightarrow NO^+ \cdot NO + NO$	$k_2 = 5 \times 10^{-30}\ cm^6\ sec^{-1}$ (15)
(R3) $NO^+(v) + N_2 + N_2 \rightarrow NO^+ \cdot N_2 + N_2$	$k_3 = 2 \times 10^{-31}\ cm^6\ sec^{-1}$ (16)
(R4) $NO^+(v) + NO \rightarrow NO + NO^+(v-1)$	k_4 = This work
(R5) $NO^+(v) + N_2 \rightarrow NO^+(v-1) + N_2^{\ddagger}$	k_5 = This work .

The experimental technique for measuring the rate k_5 was to introduce N_2 into the absorption cell described in Section 2 at increasing pressures until the recombination rate no longer dominates the disappearance of NO^+(v). Since the absorption cross section of NO^+ is constant with time, the amount of absorption is proportional to the number density of NO^+ in the lower and upper states. The decrease in NO^+(v) concentration with time is given by the sum of Reactions R1 through R5 minus the increase in NO^+(v) concentration due to Reaction R4 and R5 on the (v + 1) vibrational level. Reaction R3 becomes important when N_2 within the test chamber exceeds 30 torr in this experiment. Note that in order to measure R4 and R5 for v = 1, these reactions must be measured for v = 2, etc. However, since the initial concentration of NO^+ in v = 4 and above is negligible compared to v = 1 and v = 2, the quenching rate of v = 2 and v = 1 are not greatly effected by their

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quenching rates. Since the NO pressure was held constant at 1 torr, the effect of the association Reaction R2 was to increase the apparent rate R4 by the rate 1.6×10^{-13} cm^3 sec^{-1}.

The quenching rate of $NO^+(v = 2)$ was measured by observing the rate of extinction of light by the R2 rotational line. The overall quenching contribution to the disappearance of $NO^+(v = 2)$ is plotted in Figure 7 as a function of N_2 pressure. The pressure of NO in the system was kept constant at 1 torr. This quenching of v is the sum of the contributions of k_4 and k_5 on v minus the sum of k_4 and k_5 on $(v + 1)$. Thus,

$$k_{measured} = \frac{1}{[N_2]\tau} = k_5 + k_4 \frac{[NO]}{[N_2]} - \frac{[NO^+(v+1)]}{[NO^+(v)]} \left(k_5' + k_4' \frac{[NO]}{[N_2]} \right), \quad (9)$$

where k_4' and k_5' are the quenching rates by NO and N_2, respectively, for NO^+ in the $(v + 1)$ vibrational level. Note that the ratio of $[NO^+(v + 1)/NO^+(v)]$ is not necessarily constant with time.

The depletion rate of $NO^+(v = 3)$ was measured looking at the R12 line. However, due to the very low signal levels involved, only a very rough rate of

$$k_5(v = 3) = 2 \pm 1.5 \times 10^{-12} \text{ cm}^3 \text{ sec}^{-1} \quad (10)$$

was obtained. Since this rate was comparable to the measured rate for $v = 2$, it was assumed that the ratio of $[NO^+(v = 3)/NO^+(v = 2)]$ was constant throughout the experiment.

Substituting the value for $k_5(v = 3)$ into Eq. (8), the quenching rate of $v = 2$ gives an N_2 quenching rate of

$$k_5(v = 2) = 2.4 \pm 0.3 \times 10^{-12} \text{ cm}^3 \text{ sec}^{-1} \quad . \quad (11)$$

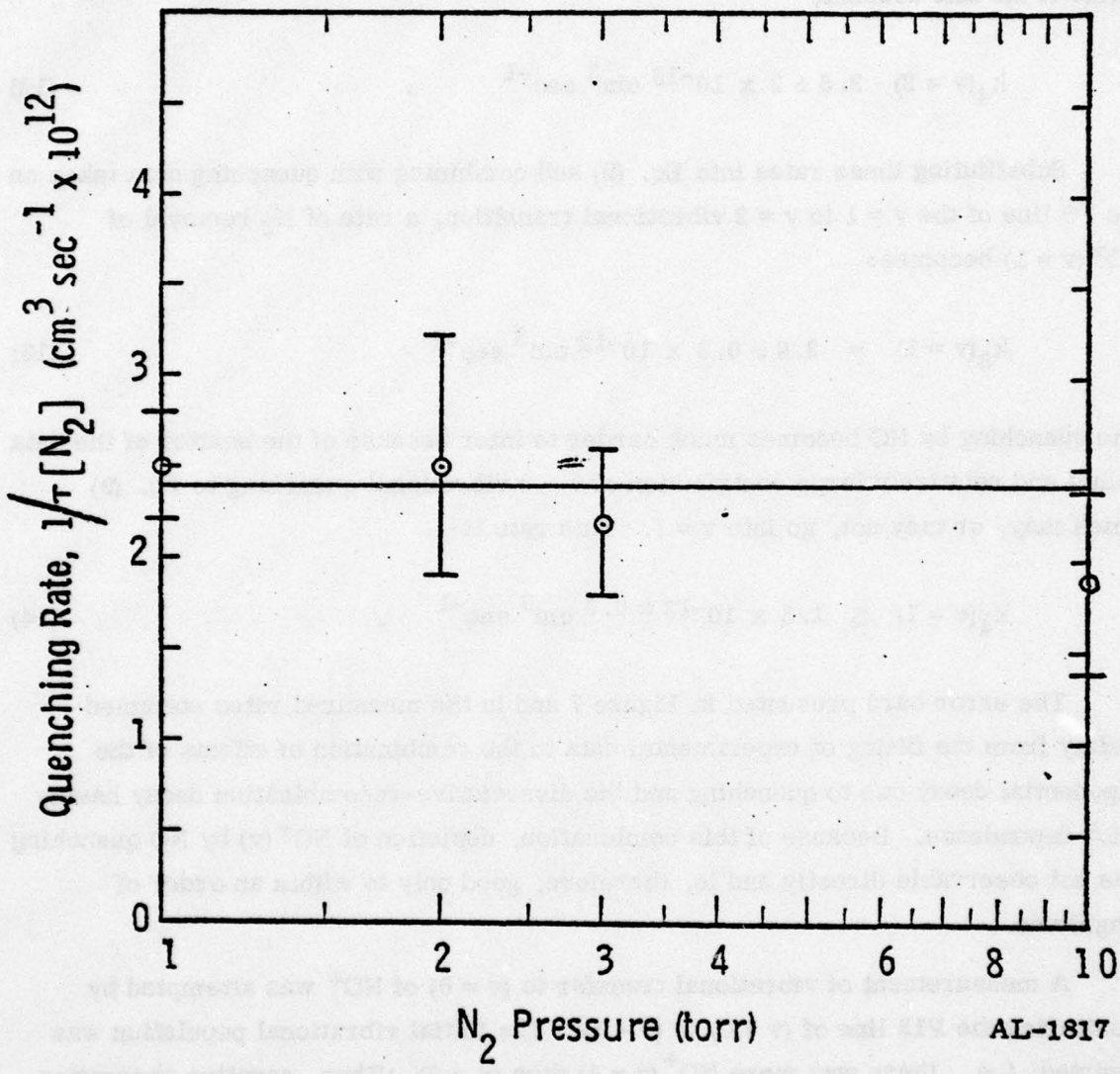

Figure 7. Quenching of v = 2 NO^+ Vibration by N_2 and NO. The Pressure of NO Was Held to 1.00 torr at All Times.

A rate for quenching by NO can also be inferred from these measurements by solving the simultaneous equations (Eq. (9)) for different N_2 concentrations and taking the effect of R2 into account;

$$k_4(v = 2) \quad 3.5 \pm 2 \times 10^{-13} \text{ cm}^3 \text{ sec}^{-1} \quad . \tag{12}$$

Substituting these rates into Eq. (9) and combining with quenching data taken on the P5 line of the v = 1 to v = 2 vibrational transition, a rate of N_2 removal of $NO^+(v = 1)$ becomes:

$$k_5(v = 1) \quad = \quad 3.0 \pm 0.5 \times 10^{-12} \text{ cm}^3 \text{ sec}^{-1} \quad . \tag{13}$$

The quenching by NO becomes much harder to infer because of the scatter of the data points and relatively large contribution of v = 2 vibrational quenching to Eq. (9) which may, or may not, go into v = 1. This rate is:

$$k_4(v = 1) \quad \lesssim \quad 1.5 \times 10^{-13 \pm 0.5} \text{ cm}^3 \text{ sec}^{-1} \quad . \tag{14}$$

The error bars presented in Figure 7 and in the measured rates stemmed mainly from the fitting of experimental data to the combination of effects of the exponential decay due to quenching and the dissociative-recombination decay having a 1/t dependence. Because of this combination, depletion of $NO^+(v)$ by NO quenching was not observable directly and is, therefore, good only to within an order of magnitude.

A measurement of vibrational transfer to (v = 0) of NO^+ was attempted by monitoring the P13 line of (v = 0) to (v = 1). The initial vibrational population was inverted, i.e., there was more $NO^+(v = 1)$ than (v = 0). Thus, negative absorption was monitored during the lamp pulse, which decayed to zero very quickly. This indicated either a more rapid recombination rate in the (v = 1) level than in (v = 0) or quenching of (v = 1) by NO in the system. Upon introduction of N_2, it was hoped that absorption would be seen at late times, when $NO^+(v = 1)$ had been quenched to

below the NO^+ (v = 0) concentration but before dissociative-recombination could take place. This effect was seen, but no quantitative information was available within the signal-to-noise of the system.

5. DISCUSSION

The radiative lifetime of NO^+ has been calculated from measurements of absorption in the v = 1 and v = 2 levels. Since these rates are a function of the relative concentration of NO^+ in the lower and upper states of the absorption, uncertainty is introduced when relating the measured values to an absorption coefficient at thermal equilibrium. From these measurements, vibrational thermal equilibrium would occur only if the majority of NO^+ vibrational excitation was quenched by N_2 before recombination could take place.

The ratio of the absorption coefficient between the v = 2 to v = 3 and the v = 1 to v = 2 transitions appear to follow the rules of a simple diatomic molecule with a linear dipole moment derivative around the equilibrium intermolecular separation, that is, $S_{lu} \sim (v + 1)\, S_{01}$ where v is the vibrational level of the lower state. Using this scaling, the integrated absorption coefficient of the v = 0 vibrational level would be 215 cm^{-2} atm^{-1} as listed in Table 2. This value is not too different from the value which Billingsley[4] would obtain using a slightly different dipole moment derivative in place of his multiconfiguration SCF calculated result. Using a single configuration Hartree-Fock calculation, Billingsley would obtain an integrated absorption coefficient of 251 cm^{-2} atm^{-1} compared to his published 88.9 cm^{-2} sec^{-1}. This value would be within 20% of our measured value. Similarly, our number differs from Michels' calculated number by 25%, well within the error bars of either our measurement or his calculations.

Our value for the integrated absorption coefficient of NO^+ in the v = 2 to v = 3 transition is not in disagreement with the Stair and Gauvin[3] observations, as their assumptions of thermodynamic equilibrium between NO^+ and CO in order to arrive at their integrated intensities, allow for errors as large as a factor of 3. Since the NO^+ produced in the upper atmosphere is by no means in thermal equilibrium and with the rapid dissociative recombination of NO^+, thermal equilibrium between NO^+ and CO would indeed be fortuitous.

The errors within this measurement of $NO^+(v)$ radiative lifetime stem primarily from determination of the concentration of $NO^+(v)$ from the dissociative recombination rate. If the recombination rate is highly dependent on the vibrational level, errors

occur not only from the determination of the time varying lower state population but also the time varying upper state population. Thus, an error in the recombination rate of NO^+ of ± 20% could change the integrated absorption coefficient as much as ± 40%. The fractional formation of NO^+ in the lower vibrational levels could also vary by ± 20% depending on the transmission of the MgF_2 windows. However, the window transmission effects mainly the v = 3 and higher levels, causing as much as a factor of 3 difference in v = 4 population. The relative concentrations of v = 1 and v = 2 levels, on the other hand, are not greatly affected by window transmission.

Other sources of error in the measurement of the NO^+ f-number are in the uncertainty within the extent of the photoionization region, which could introduce a ± 5% error and in the slight detuning of the diode laser during lamp discharge which would change the NO^+ line location by ± 0.005 cm^{-1}. These sources of error are much less important, however, than the uncertainty in the number density of $NO^+(v, J)$. Finally, the NO γ, δ, and ϵ bands excited by the UV flash lamps may all have vibration rotation lines within the NO^+ vibrational frequency range. These electronic bands have radiative lifetimes on the order of 0.4 μs, however, so their absorption of infrared radiation quickly disappears after the flash lamp turns off. The absorption of laser light by these bands during the lamp flash gave what at first appeared to be a very long spectral tail in the NO^+ absorption, which could have been prevented by using a long wavelength cut off filter in front of each flash lamp. As it turned out, because of the very short lifetime of these states, their effect was to provide a better base level from which to determine lamp shut-off.

Because 1 torr of NO was used to produce NO^+, some of the $NO^+(v)$ was quenched by NO. Thus, the instantaneous number density presented in Eq. (6) must be modified to account for this effect. Using the rates obtained in the previous section, this effect would be to increase the absorption coefficient by as much as 10%. Because of the large uncertainty in this cross section, however, this correction was not included within the analysis. While some latitude was present in varying the NO concentration within the absorption cell, a variation of a factor of 2 in NO pressure would make it black to UV radiation. Thus, the effects of NO quenching of $NO^+(v)$ could not be investigated directly using this technique.

The measurement of $NO^+(v)$ transfer to N_2 was somewhat more accurate than that of NO^+ vibrational lifetime, since the absolute number density of $NO^+(v)$ was not necessary for this determination. While the integrated absorption of excited $NO^+(v)$ scaled approximately at (+ 1), no such simple scaling was available on $NO^+(v)$ transfer to N_2. This transfer rate was seen to decrease slightly for higher vibrational levels of NO^+, rather than increasing. If this trend holds for vibrational levels higher than 3, it would be expected that NO^+ formed in the high vibrational levels would contribute most of the radiation in the upper atmosphere. Since only transfer from v = 1 and v = 2 levels were measured in detail, however, this is a premature conclusion.

While no previous measurements have been made on $NO^+(v)$ quenching by N_2, various estimates have been made based on the observations of auroral activity.[17] From the lack of detection of $NO^+(v)$ in the upper atmosphere, estimates of quenching rates as high 10^{-9} cm^{-3} sec^{-1} have been proposed. Due to the extremely good overlap of NO^+ with CO_2, however, the lack of any detectable NO^+ may be due to the fact that the signal is buried within the very large CO_2 signature. It is interesting to note that quenching of NO^+ by N_2 is on the order of 10 times more efficient than the transfer of $CO_3(\nu_3)$ to N_2. This could easily be accounted for by the ionic nature of NO^+ as well as its dipole moment. This quenching rate, together with the factor of 10^3 lower $[NO^+]$ than $[CO_2]$, even during an IBC Class II aurora,[18] could result in an NO^+ emission 10^4 weaker than $CO_2(\nu_3)$ in an auroral event.

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In conclusion, both the $NO^+(v)$ radiative lifetime and the transfer of vibration to N_2 have been measured in this experiment. While both these measurements were the first to be performed on the NO^+ ion, they agree at least qualitatively to values inferred from upper atmospheric studies. In order to obtain a better picture of the NO^+ emission during electron disturbances in the upper atmosphere, both the degree of vibrational excitation during NO^+ formation and the transfer of vibration to O and O_2 must also be measured.

6. ACKNOWLEDGMENTS

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